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理　学　硕士　学　位　论　文

环金属化钯－偶氮配合物作为光学分子传感器的应用初探

Investigation on the application of Cyclometalated Palladium-azo Complexes as Optical Molecular Chemosensors

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Investigation on the application of Cyclometalated Palladium-azo Complexes as Optical Molecular Chemosensors

A Dissertation Presented

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环金属化铂-偶氮配合物作为光学分子传感器的应用初探

摘要

光学分子传感器是“分子识别”研究在分析科学新的发展需求下的一种应用形式。由于其在环境或生物微观系统的组成、结构和衍化信息探索等方面的重要作用而日益得到重视。本论文成功地应用了一类环金属化铂-偶氮配合物作为光学分子传感器，其传感的目标物种涉及阴离子、中性有机小分子（如有机磷农药）等几类物种。论文分五章，分别包括以下主要内容：

第一章 绪论

首先简要介绍了光学分子传感器的基本概念和基本设计思路，接下来重点综述了铂族金属配合物作为光学分子传感器的研究进展，最后对这些相关研究进行分析和总结，结合本实验室的工作基础，提出本论文的研究设想。

第二章 四核环金属化铂-偶氮配合物 MOP 作为有机磷农药检测的分辨型光学分子传感器

本章利用环金属化配合物(cyclometalated complexes)的化学特性，结合我们研究组曾开发出的一类新型的分辨型光学分子传感器 MOP，考察了其对有机磷农药的选择性光谱响应能力和对不同含硫代磷酸酯骨架的有机磷农药组分的光谱分辨能力。实验表明：在 DMF－acetone 的混合溶剂中，MOP 对含硫代磷酸酯的有机磷农药有高的选择性识别能力，而对氧磷类有机磷农药（如敌敌畏）则没有光谱响应，而且 MOP 对具有不同磷酸酯骨架的硫磷类农药有较强的光谱分辨能力，籍以建立相关组分的光谱检测新方法。

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第三章 双核环金属化钯-偶氮配合物 mop 作为水溶液中硫氰酸根测定的光学分子传感器

本章利用钯-偶氮配合物 Pd-Cl-Pd 桥键的反应性能，结合铂族金属化合物对外来配体所具有的独特的分辨性光谱响应的特点，设计了水溶液中硫氰酸根的一种新型的光学分子传感器 mop。实验表明：1）在水溶液中，mop 对硫氰酸根体现出优异的识别专一性，对其它无机阴离子几乎没有响应；2）传感过程可以在较宽的 pH 范围（5.8-8.0）和高浓度的常见阴离子共存下进行，具有良好的可操作性和实用性；3）以 mop 为探针，可以在生理 pH 的水溶液中实现对硫氰酸根在 10^{-7} mol/L 水平上的定量测定。这是一种全新的硫氰酸根的光学传感理念，由于硫氰酸根阴离子在生理学和医学方面的重要性，这种光学分子传感器可望在相关的研究领域中得到借鉴和应用。

第四章 MOP 在其它生理活性物种检测方面应用的初探

本章阐述了环金属化钯-偶氮配合物在有机小分子的分辨性传感方面的应用。实验研究了 MOP 对苯胺、苯酚类化合物以及青霉素 G 的代谢产物的识别，同时探讨了其传感机理，为拓展 MOP 分子探针的在分析化学中的应用作了初步探索。

第五章 论文的总结与展望

关键词：光学分子传感器，环金属化钯-偶氮配合物，有机磷农药，硫氰酸根

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Investigation on the application of cyclometalated palladium-azo complexes as optical molecular chemosensors

Abstract

Optical molecular chemosensor (OMCS) has increasingly been important due to its distinct characteristic in probing the composition, structure and evolution of environmental or biological microsystems, and what's more, optical molecular chemosensor (OMCS) has an explosive growth in analytical chemistry. Some OMCSs have been developed in this dissertation based on a cyclometalated palladium-azo complex, where a wide range of chemical substances, including some important anions and neutral organic small molecules such as organophosphors pesticides, were explored as the sensing targets. This dissertation consists of five chapters summarized as follows.

In chapter 1, a general introduction to OMCS was presented. Emphasis was paid on the new developments of OMCS, especially those based on platinum group metal complexes. Based on reviewing the research progresses of using cyclometalated palladium-azo complexes as optical molecular chemosensors, the objective of this dissertation was presented.

In chapter 2, a tetranuclear cyclometalated palladium-azo complex (MOP) was synthesized according to our previous work and applied as a differential colorimetric molecular chemosensor for organophosphorus pesticides (OPPs) for the first time. This molecular probe was found to have highly selective

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recognition ability to OPPs. In a DMF-acetone mixed solvent, it showed no spectral response to oxophosphate pesticides, but exhibited differential absorption spectra in the presence of different sulfur phosphorus pesticides (SPPs) , which could be used for the determination of SPPs.

In chapter 3, a dinuclear cyclometalated palladium-azo complex mop was synthesized and used as an optical molecular chemosensor for thiocyanate in aqueous solution. The developed probe has the following characteristics: 1) It was found to sensitively show obvious color change upon addition of SCN^- into an aqueous solution of the probe at pH 7.0, while never show color changes upon addiction of F^-, Cl^-, Br^-, I^-, HCO_3^-, SO_4^{2-}, CH_3COO^-, et al.; 2) The absorption spectra induced by the target species hardly changed in the pH range from 5.8 to 8.0, which provides a great convenience for the feasible application; 3) The sensing of SCN^- could be performed in the presence of coexisting common anions at high concentrations; 4) SCN^- at the concentration level of 10^{-7} mol/L could be well quantitatively titrated by mop at pH 7.0. Since SCN^- is of great importance in biological chemistry, the developed probe was expected to see its application in the related research fields.

In chapter 4, MOP was further applied to sensing other bioactive targets such as phenol- or aniline-based compounds and the metabolic products of penicillin G. The sensing mechanism was also proposed. Thus attempts were made to extend the application of MOP probe in analytical chemistry.

At last, the main features and innovations of this dissertation were described.

Keywords: optical molecular chemosensors, cyclometalated

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palladium-azo complexes, organophosphorus pesticides, thiocyanate

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